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(54) ALKALINE EARTH ALUMINATE PHOSPHOR, PHOSPHOR PASTE COMPOSITION, AND VACUUM ULTRAVIOLET-EXCITABLE LUMINOUS ELEMENT

(57)Abstract:
PROBLEM TO BE SOLVED: To provide an alkaline earth aluminate phosphor which emits blue light with a high luminance under the excitation by vacuum ultraviolet (VUV) rays with wavelengths of 200 nm or less while hardly exhibiting luminance degradation; a paste composition of the same; and a vacuum ultraviolet ray- excitable luminous element which hardly suffer from the degradation in luminance and efficiently sustains the luminescence.
SOLUTION: This alkaline earth aluminate phosphor is represented by the formula: aMIIO 6Al2O3:EuX'MIIIy (wherein MII is at least one alkaline earth element selected from among Ba, Sr and Ca; MIII is at least one element selected from among Sc, Y, Gd and In; x, y and a are each a number satisfying: $0<x<1$; $0<y\leq 2$; and $0.9\leq a\leq 1.8$).

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* NOTICES *

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CLAIMS

[Claim(s)]
[Claim 1] General formula Alkaline earth aluminate fluorescent substance expressed with aMIIO and 6aluminum2O3:EuX, and MIIIy. (However, MII expresses at least a kind of alkaline-earth-metal element chosen from the group which consists of Ba, Sr, and calcium among said formula, MIII expresses at least a kind of metallic element chosen from Sc, Y, Gd, and In, and x, and y and a express the number which fulfills 0< x<1, 0< y<=2, and the conditions that become 0.9 <=a<=1.8, respectively) .
[Claim 2] The alkaline earth aluminate fluorescent substance according to claim 1 characterized by being the fluorescent substance for vacuum-ultraviolet-radiation excitation with which said fluorescent substance emits light under vacuum-ultraviolet-radiation excitation with a wavelength of 200nm or less.
[Claim 3] The fluorescent substance paste constituent characterized by said fluorescent substance being an alkaline earth aluminate fluorescent substance according to claim 1 or 2 in the fluorescent substance paste constituent which comes to carry out distributed content of the fluorescent substance into the solvent which dissolved the binder.
[Claim 4] The fluorescent substance paste constituent according to claim 3 characterized by the content of said fluorescent substance being 5 - 70 % of the weight.
[Claim 5] The vacuum-ultraviolet-radiation excitation light emitting device characterized by using an alkaline earth aluminate fluorescent substance according to claim 1 or 2 for said fluorescent screen in the vacuum-ultraviolet-radiation excitation light emitting device which excites said fluorescent screen with the vacuum ultraviolet radiation emitted by discharge of the rare gas enclosed in the vacuum envelope by which the fluorescent screen was formed in the interior, and is made to emit light.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] Especially this invention relates to the vacuum-ultraviolet-radietion (VUV) excitation light emitting device which brightness degredation may make meintain luminescence efficient few at the alkaline earth aluminate fluorescent substance with which wavelength presents little blue luminescence of high brightness of brightness degredation under excitation by vacuum ultraviolet radiation (VUV) 200nm or less, and the fluorescent substance paste constituent list which used this fluorescent substance.
[0002]
[Description of the Prior Art] For example, so that it may be represented by a rare-gas lamp, a plesma display panel (PDP), etc. which are used for the light source for read of a scanner While forming the fluorescent screen which used the fluorescent substance which emits light under excitation by VUV in the envelope which consists of glass etc. Or it mixes and encloses. the inside of it — rare gas, such as Ar, Xe, helium, end Ne, — a simple substance — Development of a VUV excitation light emitting device with the structure and the function to excite the fluorescent screen in an envelope and to make it emit light by VUV emitted by making the enclosed rare gas discharge is performed briskly in recent years, and is used.
[0003] As a fluorescent substance conventionelly used es e fluorescent screen of this VUV excitation light emitting device, it is BO (Y, Gd)3. : Red fluorescent substances, such as Eu, LaPO4: — green luminescence fluorescent substances, such as Ce, Tb, Zn2SiO4:Mn, BaAl12O19:Mn, O-aAl(Ba, Sr, Mg) 2O3:Mn, and YBO3:Tb, — BeMgAl10O17:Eu and MgAl(Ba, Sr) 10O17: -- blue luminescence fluorescent substances, such as Eu and Mn, etc. — a single — or it is mixed and used.
[0004] As e property of the fluorescent substance used as a fluorescent screen of a VUV excitation light emitting device What (there is little brightness degredation by baking) the luminescence brightness as a fluorescent screen does not fall in case a fluorescent substance peint film receives the beking processing around 500 degrees C with the fluorescent screen formation process of emitting light in high brightness more under excitation by VUV, and e VUV excitation light emitting device, Although it is required that a fluorescent substance has few brightness falls (brightness degradation by VUV), that the color purity of the luminescent color should be good, etc. even if it carries out long duration octuetion of the VUV excitation light emitting device end is continuously exposed to VUV, the fluorescent substance by which current utilizetion is carried out does not satisfy all of these properties, either. On the other hand, in the commercial scene, there is always much more improvement demand of many properties of a VUV excitation light emitting device, and development of a new fluorescent substance with said property good also about the fluorescent substance for VUV excitation is expected. By the wey, aluminate fluorescent substances are typical blue thru/or a fluorescent substance for VUV excitation of bluish green color luminescence in the fluorescent substance for VUV excitation. BeMgAl10O17:Eu and MgAl(Be, Sr) 10O17: — Eu, Mn, etc. The fluorescent substance called the common-name BAM fluorescent substance which used divalent Eu, or Eu and Mn es the activator to the aluminate of the alkaline earth metal which contains Mg as indispensable as host

crystal is used as the blue for VUV excitation excellent in luminescence properties, such as luminescence brightness, thru/or a bluish green color luminescence fluorescent substance. However, especially this BAM fluorescent substance is a fluorescent substance with which brightness degradation by VUV had a large fault in the brightness degradation list by baking, and development of blue with little brightness degradation by baking which replaces this and brightness degradation by VUV thru/or the fluorescent substance for VUV excitation of bluish green color luminescence is desired.
[0005] The effectiveness that the alkaline earth aluminate fluorescent substance expressed with general formula a(M1-x Eux) O and 6aluminum 2O3 which this invention persons proposed previously has little brightness degradation by high brightness and VUV as a blue fluorescent substance for VUV excitation is done so (refer to application for patent No. 245132 [2001 to]). However, to reduce practically extent of brightness degradation by VUV of this alkaline earth aluminate fluorescent substance further is desired.
[0006]
[Problem(s) to be Solved by the Invention] This invention is made in order to conquer the trouble of the conventional technique according to the aforementioned request, and luminous efficiency is high, there is especielly little brightness degradation by VUV es a fluorescent substance for VUV excitation, and it aims at providing with a VUV excitation light emitting device the alkaline earth aluminete fluorescent substance which presents blue luminescence of high brightness, and the fluorescent substance paste constituent list using this fluorescent substance.
[0007]
[Means for Solving the Problem] Then, this invention person etc. reaches [that brightness degradation by VUV excitation fells further, and] a header and this invention by making this fluorescent substance contain a specific metallic element, as e result of continuing about the fluorescent substance expressed with general formula a(M1-x Eux) O and 6aluminum 2O3 proposed by said epplication-for-petent No. 245132 [2001 to] specification and adding examination wholeheertedly.
[0008] That is, if this invention is summarized, it will consist of each following invention and a mode.
(1) General formula Alkaline earth aluminate fluorescent substance expressed with aMIIO and 6aluminum2O3:EuX, and MIIIy. (However, MII expresses at least e kind of alkaline-earth-metal element chosen from the group which consists of Ba, Sr, and calcium among said formula, MIII expresses at least a kind of metallic element chosen from Sc, Y, Gd, and In, end x, and y and a express the number which fulfills 0< x<1, 0< y<=2, and the conditions that become 0.9 <=a<=1.8, respectively) .
(2) said — y — e value — x — a value — and — a — a value — respectively — 0.03 — <= — x — <= — zero . — five — zero — < — y — <= — one — and — 1.1 — <= — a — <= — 1.5 - - it is — things — the description — ** — carrying out — the above — (— one —) — a publication — an alkaline earth — an aluminate — a fluorescent substance .
(3) Said MII is the above (1) characterized by consisting of Ba beyond 50 mol % of said alkaline-earth-metal element, or an alkaline earth aluminate fluorescent substance given in (2).
[0009] (4) An alkaline earth aluminete fluorescent substance given in any of aforementioned (1) - (3) characterized by being the fluorescent substance for vacuum-ultraviolet-radiation excitation with which said fluorescent substance emits light under vacuum-ultraviolet-radiation excitation with e wavelength of 200nm or less they are.
(5) The fluorescent substance paste constituent characterized by this fluorescent substance being an alkaline earth eluminate fluorescent substance given in any of eforementioned (1) - (4) they are in the fluorescent substance peste constituent which comes to carry out distributed content of the fluorescent substance into the solvent which dissolved the binder (binder).
[0010] (6) A fluorescent substance paste constituent given in the above (5) characterized by the content of seid fluorescent substance being 5 - 70 % of the weight.
(7) The vecuum-ultraviolet-radietion excitation light emitting device cheracterized by using an alkaline earth aluminete fluorescent substance given in any of aforementioned (1) - (4) they are

for this fluorescent screen in the vacuum-ultraviolet-radiation excitation light emitting device which excites this fluorescent screen with the vacuum ultraviolet radiation emitted by discharge of the rare gas enclosed in the vacuum envelope by which the fluorescent screen wes formed in the interior, and is made to emit light. In addition, the fluorescent substance expressed with general formula aMIIO and 6aluminum2O3:EuX, and MIIIy in this invention means the fluorescent substance with which the percentage of each metallic element of MII, aluminum and Eu which are contained in the fluorescent substance, and MIII is satisfied of the above-mentioned general formula.
[0011]
[Embodiment of the Invention] Hereefter, this invention is explained to a detail. The alkaline earth aluminate fluorescent substance of this invention 1) At least one sort of alkaline earth elements chosen from the group which consists of Ba, Sr, and calcium, 2) Each oxide of Eu element which are at least a kind of element chosen from the group which consists of aluminum element, 3Sc, and Y, Gd and In, and four activator, Or compounds, such as a carbonete of each element of these 1-4, a sulfate, and a halogenide To stoichiometric, aMIIO and 6aluminum2O3:EuX, MIIIy (However, MII expresses at least one sort of alkaline-earth-metal elements chosen from the group which consists of Ba, Sr, and celcium among said formula.) MIII expresses at leest a kind of metallic element chosen from Sc, Y, Gd, and In, end x, and y end a express the number which fulfills 0< x<1, 0< y<=2, and the conditions that become 0.9 <=a<=1.8, respectively. It is the same as thet of the following. **** at a becoming rate and the fluorescent substance raw material compound which consists of such mixture is mixed enough. It distributes like the tail end process which fills up heat-resistant containers, such as alumina crucible, celcinates, and is applied to the obtained baking object at the time of the usual fluorescent substance manufacture. The alkaline earth aluminete fluorescent substance of this invention expressed with general formula aMIIO and 6aluminum2O3:EuX, and MIIIy can be manufactured by performing rinsing, desiccation, and many processings of sieving. In addition, the fluorescent substance expressed with general formula aMIIO and 6aluminum2O3:EuX, and MIIIy in this invention means the fluorescent substance with which the percentage of each metallic element of MII, aluminum and Eu which are contained in the fluorescent substance, and MIII is satisfied of the above-mentioned general formula. In the fluorescent substance raw material compound with which baking is presented, it can also add in the mixture of a fluorescent substance raw materiel compound by making fluorides, such as AlF3, BaF2, and HF (NH4)2, into flux for promotion of e reaction as well as the cese of the conventional alkaline earth eluminate fluorescent substance manufactures, such as a BAM fluorescent substance.
[0012] Although the content (x values) of Eu used es an ectivator of it being necessary to be size is more natural than 0 in order to obtain the fluorescent substance which may emit light blue under a certain stimulus in this invention since concentration quenching is started and it stops almost emitting light, when x velues exceed 1 — x velues — 0 — size — 1 — smallness - - it is necessary to carry out — the content (y value) of e MIII element — 2 — size — luminescence — ** and beem luminescence brightness fall remarkably. Moreover, about e value, it is smaller than 0.9, or since the fluorescent substance which whose amount of mixture of an impurity component increased in chemical composition when it was size, and excelled [brightness / high] 1.8 in VUV-proof nature is not obtained, it is not desirable. Therefore, the alkeline earth aluminate fluorescent substance of this invention In respect of luminescence brightness, the content (x values) of Eu and the content (y value) of a MIII element From the viewpoint of luminescence brightness when considering as the presentation set to 0< x<1 and 0< y<=2 in respect of luminescence brightness, respectively cerries out VUV excitation preferably, it is more desirable especially that x values, y value, and a value are 0.03<=x<=0.5, 0< y<=1, and 1.1<=a<=1.5, respectively. It is in the inclination for the luminescence brightness under VUV excitation of the fluorescent substance obtained even if x values are smeller than 0.03 and it exceeds 0.5 to become low. moreover, the luminescence brightness under VUV excitation of the fluorescent substance which will be obtained if y value exceeds 1 — ** — it becomes low. Moreover, about a value, it is smaller than 1.1, or since it becomes difficult to obtain the fluorescent substance which mixture of an impurity component arose in chemical composition

when it was size, and excelled [brightness / high] 1.5 in VUV-proof nature a little, it is not so desirable. As for the alkaline earth aluminate fluorescent substance of the viewpoint of the luminescence brightness (resultant stimulus) when carrying out VUV excitation to this invention, it is desirable thet M element which constitutes some host crystal of a fluorescent substance is Be, or they are less than [50 mol %] and the alkaline-earth-metal element which permuted Be not more then 20 mol % by at leest one in Sr and calcium more preferably.
[0013] A fluorescent substance raw material compound is calcinated once or more over 2 - 40 hours in a reducing atmosphere according to the fill at the temperature of 1300-1800 degrees C. The fluorescent substance of this invention like the alkaline earth aluminate fluorescent substance which does not contain the MIII element of a publication on application-for-patent No. 245132 [2001 to] specifications Although it hes the property thet a broad peak is observed in the include-angle field of 28-31 degrees of angle of diffractions in the powder diffraction X-ray spectrum by the CuKelpha1 characteristic X ray of this fluorescent substance (2theta), and little brightness degradation by VUV is If burning temperature is made lower than 1300 degrees C, the above-mentioned broad peak in the powder diffraction X-ray spectrum of the fluorescent substance obtained will no longer be accepted gradually. If sufficient luminescence brightness under VUV excitation is not obtained when extent of brightness degradation by VUV also becomes large with it, and it is made higher than 1800 degrees C, unnecessary energy will be consumed and it is not industrially desirable. Moreover, in order to acquire the reducing etmosphere at the time of baking, the approach of embedding a graphite end ectivated carbon into the crucible with which the fluorescent substance raw material compound wes filled up, the approach of embedding the crucible filled up with the fluorescent substance raw material compound in the crucible filled up with a graphite or activated carbon, the epproach of calcinating in the mixture of gas of nitrogen end hydrogen, etc. are mentioned. Furthermore, the steam may be contained in the firing environments.
[0014] Next, the fluorescent substance paste constituent of this invention is described. The fluorescent substance paste constituent of this invention contains the component currently used in the conventional fluorescent substance peste constituent except using the alkeline earth aluminate fluorescent substance of this invention which is the above as fluorescent substance powder, and wes made end obtained. The fluorescent substance peste constituent of this invention is manufactured like the case where the conventional fluorescent substance paste constituent is manufactured, except using the alkaline earth aluminate fluorescent substance of this invention. For example, while fully agitating and kneading the mixture which carried out specified quantity mixing of the elkaline eerth aluminate fluorescent substance of this invention, and the solvent in which binder resin was dissolved, respectively and distributing a fluorescent substance, it can obtain by adjusting to the viscosity which suited the purpose of use.
[0015] In order to use ethyl cellulose, a nitrocellulose, polyethylene oxide, acrylic resin, etc. according to the purpose of use on the occasion of manufacture of the fluorescent substance paste constituent of this invention as binder resin used with said alkaline earth aluminate fluorescent substance and to distribute a fluorescent substance and binder resin, as a solvent used with a fluorescent substance and binder resin for viscosity control, water, butyl acetate, butyl carbitol, butyl carbitol acetate, the Tell Young Pioneers, etc. are mentioned. As for the loadings of an alkaline earth aluminate fluorescent substance, it is desirable to consider as 5 - 70 % of the weight to the total weight of the fluorescent substance and binder resin except a solvent, to mix with the solvent which dissolved binder resin with this fluorescent substance, to agitate and kneed this, and to add and carry out viscosity control of the solvent finally in respect of control of film thickness, the workability of spreading, etc.
[0016] Next, the VUV excitation light emitting device of this invention is explained in full detail. When manufacturing the rare-ges lamp which is one of the VUV excitation light emitting devices of this invention For example, flow coating of the fluorescent substance paste constituent of this invention which had viscosity adjusted even from the end of the transparent glass cepillary which has e desired bore to extent which can flow a tubing internal-surface top is carried out.
[whether this is dried, baking processing is carried out further, end baking vaporization of the organic substance component is carried out, and] Or after putting the glass plate which carried

out spreading desiccation of the fluorescent substance paste constituent into the interior of a glass capillary. After exhausting the interior of the glass tube, in tubing, a small amount of rare gas is enclosed, an electrode is attached in both sides to which inside-and-outside both sides which sandwich the both ends or glass tube wall of a glass capillary, or the exterior of a glass tube countered, and the both ends of the glass tube are stopped. Thus, it considers as the rare-gas lamp which is one of the VUV excitation light emitting devices of this invention.

[0017] Moreover, when manufacturing PDP which is other one example of the VUV excitation light emitting device of this invention, an internal electrode is formed in tooth-back plates, such as a glass plate, the septum of the shape of the shape of a stripe and a matrix is formed, two or more cels are constituted, and red, green, and a blue fluorescent substance paste constituent are applied to a wall by approaches, such as screen printing, at red, green, and the pars-basilaris-ossis-occipitalis list of each septum which constitutes a cel for every blue color. The fluorescent substance paste constituent of this invention is used as a blue fluorescent substance paste. While drying and baking this and forming a fluorescent screen in each cel, after carrying out opposite arrangement of the front plate which consists of a glass plate with which a tooth-back plate and fixed spacing were separated, and the internal electrode was formed, stopping the perimeter of a front plate and a tooth-back plate and exhausting the interior, rare gas is enclosed and it is referred to as PDP which is one of the VUV excitation light emitting devices of this invention.

[0018] In addition, outside the aforementioned rare-gas lamp or PDP, the VUV excitation light emitting device of this invention encloses rare gas in the envelope in which the fluorescent substance paste constituent of this invention was applied to the front face of the base material used as the luminescence side in an envelope [in / not related / how / gestalt / the class / each VUV excitation light emitting device] by the well-known approach, this was dried, baking processing was carried out, each fluorescent screen was formed in, and the fluorescent screen was formed, and is manufactured. Thus, the VUV excitation light emitting device of obtained this invention can obtain the VUV excitation light emitting device of high brightness with few falls of working luminescence brightness.

[0019]

[Example] Next, this invention is not limited by the following examples, although the example of this invention is given with the example of a comparison and this invention is explained concretely.

[0020] [Example 1A]

a BaCO3molSc2O3 :0.08 molAlF3 :0.05 mol -- after mixing said each fluorescent substance raw material enough, it calcinated over 24 hours including rising-and-falling-temperature time amount at 1600 degrees C of maximum temperatures in the nitrogen which filled up alumina crucible, put in the graphite, covered and contained the steam. : 1.1574 Mol Eu 2O3 : 0.0643 Mol aluminum 2O3 : 5.94 Subsequently, baking powder was sifted and the alkaline earth aluminate fluorescent substance of example 1A was obtained.

[0021] [Example 1B] 30% of the weight of the alkaline earth aluminate fluorescent substance of example 1A, 10% of the weight of butyl carbitol, 53% of the weight of butyl carbitol acetate, and 7% of the weight of ethyl cellulose were fully kneaded, and the fluorescent substance paste constituent of example 1B was manufactured.

[0022] The fluorescent substance paste constituent of example 1B which is the [example 1C] above, and was made and obtained was applied on the glass plate with a width of face of 2mm, and was calcinated at 500 degrees C after 60-minute desiccation by 120 degrees C for 30 minutes. After having held this glass plate in the glass tube with an outer diameter of 4mm, attaching the electrode of nickel to the both ends of this glass tube and exhausting the inside of tubing to a vacuum, 50Torr enclosure of Ne98%-Xe2% of the gas was carried out, and the VUV excitation light emitting device (rare-gas lamp) of example 1C was produced. When [which carries out continuation lighting of the VUV excitation light emitting device of this example 1C, asks for the resultant stimulus (brightness/y) 96 hours after switching on the light in the list immediately after lighting, respectively, and receives immediately after lighting] the relative value (resultant-stimulus maintenance factor) of the resultant stimulus (brightness/y) 96 hours

after switching on the light was computed, the resultant-stimulus maintenance factor of the VUV excitation light emitting device of example 1C was 99.1%. In addition, although the brightness of a blue luminescence fluorescent substance changes a lot in proportion to the luminescent color (y value of a chromaticity point), generally comparing with the value which considered as the simple approach of carrying out the mutual comparison of the luminous efficiency between the fluorescent substances with which the luminescent color (y value) differs, and broke brightness by y value is performed. Then, also in this invention, the measured value of luminescence brightness carried out the relative comparison mutually in quest of the resultant stimulus (brightness/y) of said definition, respectively.

[0023] The compounding ratio of the fluorescent substance raw material which expressed with the percentage of each metallic element contained in the fluorescent substance raw material used when the fluorescent substance of example 1A was manufactured to Table 1, and the resultant-stimulus maintenance factor of the VUV excitation light emitting device of example 1C are shown, respectively.

[0024] The fluorescent substance of example of comparison 1A was obtained in example 2A - 4A list like example 1A except having used the fluorescent substance raw material of the compounding ratio shown in Examples 2A-4A and the [example of comparison 1A] table 1.

[0025] The fluorescent substance paste constituent of example 2B-4B and example of comparison 1B as well as the fluorescent substance paste constituent of example 1B was manufactured except having replaced with the fluorescent substance of [example 2B-4B and example of comparison 1B] example 1A, and having used each fluorescent substance of Examples 2A-4A and example of comparison 1A.

[0026] It is made to be the same as that of the VUV excitation light emitting device of example 1C except having replaced with the fluorescent substance paste constituent of ExamplesC [2]-4C and [example of comparison 1C] example 1B, and having created the fluorescent screen using each fluorescent substance paste constituent of example 2B-4B and example of comparison 1B. The VUV excitation light emitting device of Examples 2C-4C and example of comparison 1C is manufactured, and the result of having measured the resultant-stimulus maintenance factor of this VUV excitation light emitting device like example 1C is shown in Table 1.

[0027]

[Table 1]

	Ba元素／モル	Eu元素／モル	Al元素／モル	III元素種類	III元素／モル	初期輝度維持率 (%)
実施例1	1.1574	0.1286	11.88	Sc	0.12	99.1
実施例2	1.1574	0.1286	11.88	Y	0.12	97.0
実施例3	1.1574	0.1286	11.88	Gd	0.12	98.6
実施例4	1.1574	0.1286	11.88	In	0.12	96.5
比較例1	1.1574	0.1286	12	-	0	94.1

[0028] As shown in Table 1, compared with the VUV excitation light emitting device of example of comparison 1C using the alkaline earth aluminate fluorescent substance with which the VUV excitation light emitting device (examples 1C-4C) which makes a fluorescent screen the alkaline earth aluminate fluorescent substance of this invention which contains a MIII element during a presentation does not contain a MIII element during a presentation, extent of brightness degradation by VUV of a fluorescent substance was reduced, and the resultant-stimulus maintenance factor has been improved remarkably.

[0029]

[Effect of the Invention] Eu activation alkaline earth aluminate fluorescent substance of this

invention and the fluorescent substance paste constituent using this present efficient blue luminescence by VUV excitation with a wavelength of 200nm or less, and since there is little brightness degradation by VUV, it becomes possible [the VUV excitation light emitting device manufactured using the fluorescent substance paste constituent of this invention] for change of the luminous efficiency under long duration actuation to maintain little luminescence of high brightness.

[Translation done.]